ROBL-CRG	**Experiment title:** EXAFS investigations of U(VI) uptake by cementitious materials	**Experiment number**: 20-01-674
Beamline: BM 20	**Date of experiment**: from: 14/04/2008 at 8:00 to: 18/04/2008 at 8:00	**Date of report**: 14.11.08
Shifts: 9	**Local contact(s)**: Dr. Dipanjan BANERJEE (banerjee@esrf.fr)	*Received at ROBL:*

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Uranium L_{III}-edge (17166 eV) extended X-ray absorption fine structure (EXAFS) spectra were recorded at room temperature (RT). We used XAS spectroscopy to determine the local chemical environment of U(VI) in cementitious materials: calcium silicate hydrates (C-S-H phases) and crushed and fully hydrated hardened cement pastes (HCP). In addition, Soddyite, Uranophane and $CaUO_4.H_2O$ (phase X) have been measured as U(VI) references compounds. EXAFS data were reduced with the Iffefit software package. Theoretical scattering paths were calculated with FEFF8.0, including multiple scattering paths.

Figure 1a shows EXAFS spectra for the most important U(VI) reference compounds ($CaUO_4.xH_2O$ or phase X, uranophane and soddyite), employed as dry powder, together with those of U(VI) sorption samples (HCP/U(VI) and C-S-H/U(VI)), which were used as wet pastes. **Figure 1b** shows their corresponding Radial Structural functions (RSFs) calculated from Fourier transforming k^3-weighted EXAFS functions between 2 and 11 Å. The sorption samples were prepared by adding U(VI) solution to crushed-fully-hydrated HCP in ACW or C-S-H suspension, respectively, and subsequently equilibrating the suspensions for 30 days. After separation from the supernatant solution by centrifugation, the wet pastes were mounted in a Plexiglas sample-holder and sealed with kapton® tape.

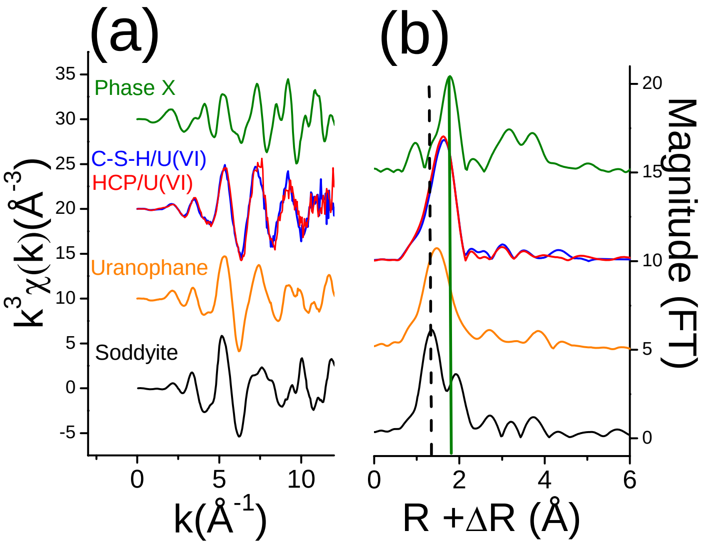

Figure 1: k^3-weighted experimental EXAFS data for selected U(VI) reference compounds and U(VI) sorption samples (a) and their corresponding RSFs (b), k range was taken between 2 and 11 Å^{-1} for all FTs.

At low loadings, the EXAFS data showed that the chemical environments of U(VI) taken-up by C-S-H and HCP are similar. Thus, one may conclude that C-S-H phases are most likely responsible for the U(VI) immobilization in HCP. Furthermore, at low loadings, the chemical environments of the U(VI) taken-up by C-S-H and HCP species are different from a soddyite-like or phase-X-like coordination environment but resemble that of U(VI) in uranophane.

Future Outlook:

The next step of this study is to determine the long term behavior of U(VI) in cementitious systems. The next XAS measurements will be dedicaced to U(VI) immobilized samples in C-S-H (with variable C/S ratio) and HCP with at least 300 days of contact time. The measurements will be done at RT.

Conferences contributions:

07/2008 : Actinide XAS 2008, 5th workshop on speciation, techniques and facilities for radioactive materials at synchrotron light sources (Saint-Aubin, France). **N. MACE, M. HARFOUCHE, R. DÄHN, J. TITS, A. SCHEINOST AND E. WIELAND**. *EXAFS Investigation of U(VI) Speciation In Cementitious Materials*. **Poster**